

## *Streak Camera Records Ultrafast Lattice Dynamics*

**X** rays have long been used as structural probes of complex molecules and solids on the atomic scale, but only recently have these techniques been extended into the time domain. At the Advanced Light Source (ALS), an international collaboration led by researchers from the University of California, Berkeley, developed and then applied a high-speed x-ray streak camera to watch—in real time—the motion of atoms in the semiconductor indium antimonide on picosecond time scales. Following a “kick” from a short laser pulse, they directly observed large-amplitude coherent atomic vibrations, in which the atoms collectively oscillated about their equilibrium positions. At higher laser powers, they were able to follow the structural phase transition from an ordered to a disordered state in real time.

The investigators performed their experiment on Beamline 7.3.3. A femtosecond laser pulse synchronized to the individual

electron bunches in the storage ring with a jitter of less than 5 picoseconds was made to overlap—in both space and time—a single x-ray pulse on the crystal. A monochromator selected x rays of wavelength 2.4 Å that illuminated a crystal oriented to diffract from the (111) planes onto the detector, a streak camera with 3-picosecond resolution. They then followed the structural dynamics of the crystal by monitoring the intensity of the diffracted x rays as a function of time with the camera. Since the length of the x-ray pulse was hundreds of times longer than the laser pulse, it served as a “continuous” source of x rays for diffraction before, during, and after the laser excitation of the sample.

Impulsive laser excitation of the crystal initially created “hot” electrons whose energy quickly was transferred to lattice vibrations, thereby heating the crystal. Following the excitation, the intensity of diffracted x rays decreased as expected, owing to a

shift in the wavelength of the Bragg peak as the heated lattice expanded. But following the decrease, the team observed distinct temporal oscillations in the diffracted intensity, indicative of coherent lattice motion, in contrast to the incoherently excited lattice vibrations of a crystal in thermal equilibrium. By slightly changing the angle of the crystal with respect to the incident x rays, different vibrational modes, or phonons, could be selected, thus mapping out part of the acoustic phonon dispersion relation. This new technique thus allows one to probe the vibrational properties of solids at frequencies up to 0.1 THz, even under extreme, highly nonequilibrium conditions, by directly watching the atoms collectively ring.

Because the investigators were able to resolve in real time the transfer of energy from the carrier system to the lattice, important physical parameters such as the electron–acoustic phonon coupling time could be extracted

by quantitatively fitting the data to models of the processes involved. In addition, the researchers found that a significant contribution to the excitation of the coherent phonon state was also due to a direct coupling between the carriers and the acoustic phonons through the deformation potential interaction.

Furthermore, the team found a close relationship between the excitation of this coherent phonon state and the disordering transition that occurred above a critical laser fluence. In particular, the diffracted x-ray signal disappeared on a time-scale determined by a vibrational period, implying that each mode took one final collective swing in one direction before disordering. Further time-resolved observations of phase transitions in other materials (for example, strongly correlated systems) should lead to greater understanding of the driving mechanisms behind them.

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A.M. Lindenberg, I. Kang, S.L. Johnson, T. Misalla, P.A. Heimann, Z. Chang, J. Larsson, P.H. Bucksbaum, H.C. Kapteyn, H.A. Padmore, R.W. Lee, J.S. Wark, and R.W. Falcone, “Time-Resolved X-Ray Diffraction from Coherent Phonons during a Laser-Induced Phase Transition,” *Phys. Rev. Lett.* **111** (2000).

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# X-RAY DIFFRACTION FROM COHERENT PHONONS

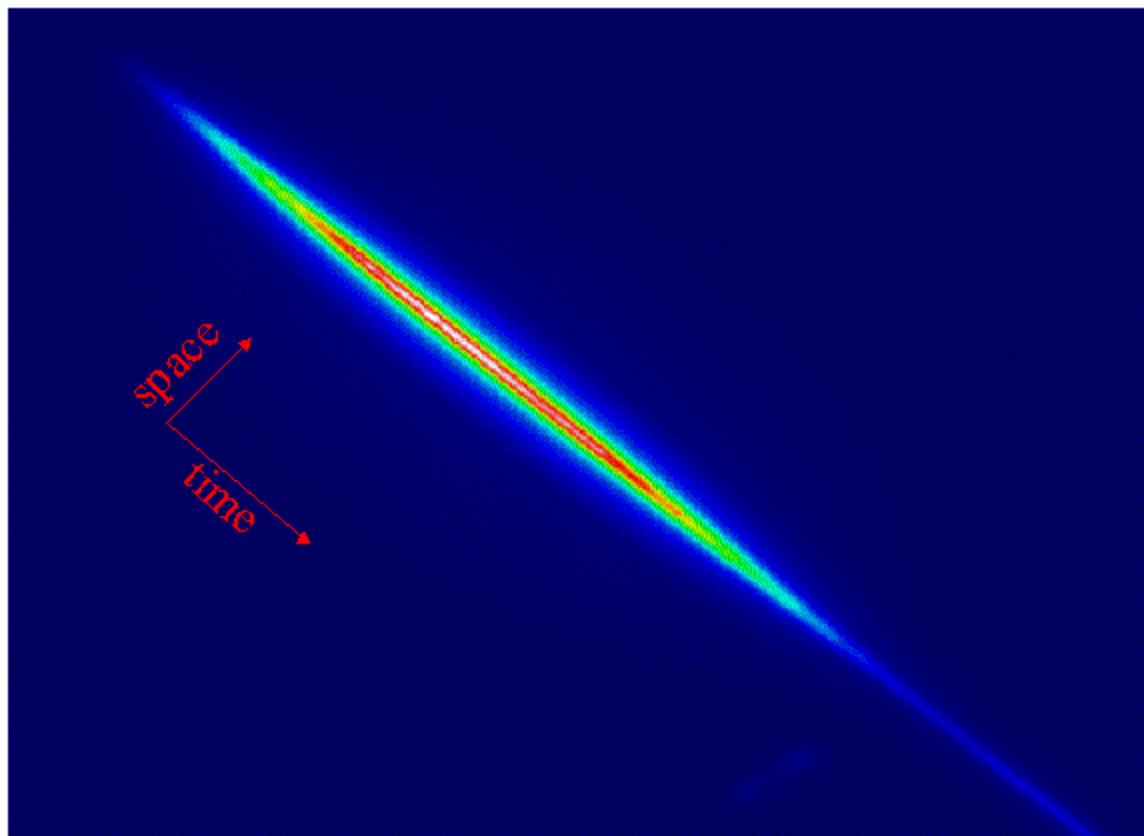


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- **Time-resolved x-ray structural probes at atomic level**
  - *New extension into time domain of static techniques*
  - *Track dynamical processes in solids, complex molecules*
- **Streak camera provides ultrafast time resolution**
  - *Femtosecond laser excites the process under investigation*
  - *Camera records data in real time with “continuous” x-ray probe*
  - *Limited by speed of camera to about 1 picosecond*
- **Coherent phonons observed in laser-excited InSb**
  - *Map acoustic phonon dispersion relation*
  - *Extract physical parameters like electron-phonon coupling time*
  - *Detect additional direct deformation potential contribution*
- **Disorder transition above a critical laser fluence**
  - *Occurs in time determined by the vibrational period*
  - *Lattice makes one collective “ring” before disordering*

# X-RAY DIFFRACTION FROM COHERENT PHONONS

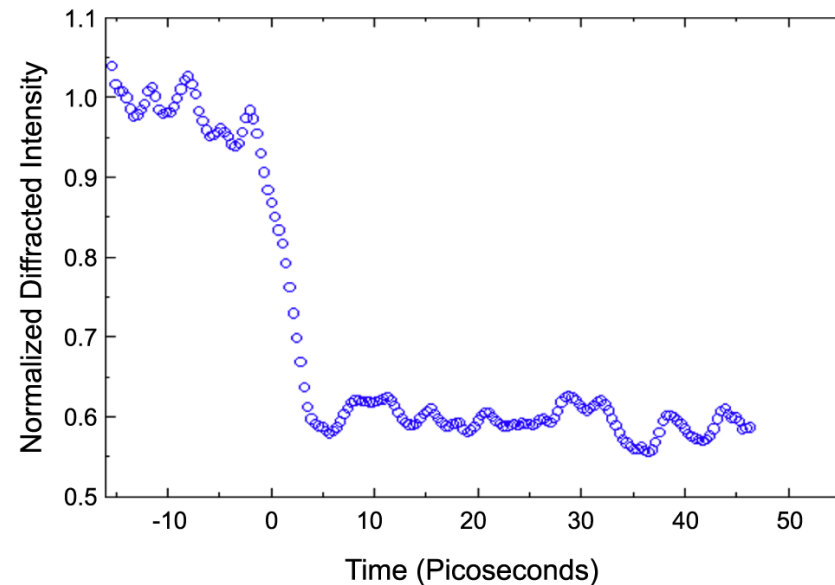
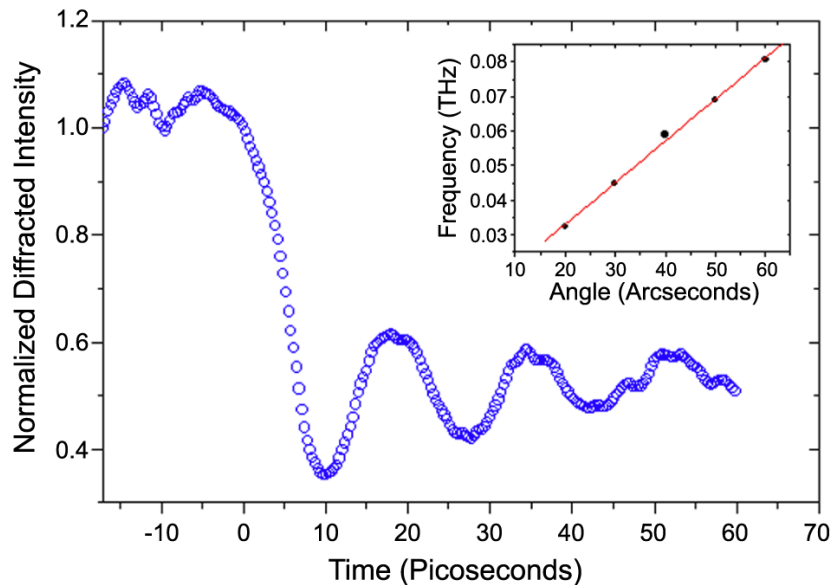
## *Streak Camera Records Ultrafast Lattice Dynamics*



*Streak camera image of a single 60-picosecond x-ray pulse. Time runs from the upper left to the lower right*

# X-RAY DIFFRACTION FROM COHERENT PHONONS

## Streak Camera Records Ultrafast Lattice Dynamics



*(left) The oscillatory signal following laser excitation is indicative of coherent, large-amplitude lattice vibrations whose frequency spectrum can be found by changing the diffraction angle relative to the Bragg peak (inset).*

*(right) Above a critical laser fluence, the sample is coherently driven into a disordered state.*